

IN THE U.S. PATENT AND TRADEMARK OFFICE

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In re application of : Before the Board of Appeals

Koshiro SHIMAZU et al. : Appeal No.:

Serial No.: 09/197,499 : Group No.: 1754

Filed: November 23, 1998 : Examiner: NGUYEN

For: RANEY CATALYST, PROCESS FOR PRODUCING IT AND
PROCESS FOR PRODUCING A SUGAR-ALCOHOL USING THE
SAME



November 8, 2002
Docket No.: 1984-0105P

APPEAL BRIEF

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Honorable Commissioner of
Patents and Trademarks
Washington, D.C. 20231

November 8, 2002
Docket No.: 1984-0105P

Sir:

CH-13038
This Appeal Brief is respectfully submitted on behalf of the
Appellants in connection with the above-identified application.

This is an appeal from the Examiner's Final Office Action
dated December 18, 2001 finally rejecting claims in connection
with the present application. The appealed claims are presented
in the attached Appendix.

REAL PARTY IN INTEREST

In accordance with 37 C.F.R. 1.192(c)(1), it is submitted
that the real party in interest with respect to the present
application constitutes the Assignees of the present application:

1) TOWA CHEMICAL INDUSTRY, CO., LTD.; and 2) NIKKO RICA CORPORATION, both of Tokyo, Japan.

RELATED APPEALS AND INTERFERENCES

In accordance with 37 C.F.R. 1.192(c)(2), it is submitted there are no other appeals or interferences known to the appellants, the undersigned, or the Assignees which will directly affect or be directly affected by or have a bearing on the Board's decision in the present appeal.

STATUS OF THE CLAIMS ON APPEAL

Claims 1, 18 and 20 are appealed from the Examiner's Final Office Action dated December 18, 2001, finally rejecting these claims. Claims 11-17 are withdrawn from consideration as being drawn to nonelected subject matter.

STATUS OF AMENDMENTS

The Amendment under 37 CFR 1.116 was filed July 12, 2002. The Examiner indicated on the Advisory Action (paper #22) that the July 12, 2002 Amendment will be entered upon Appellants filing of a Notice of Appeal and an Appeal Brief.

SUMMARY OF THE INVENTION

There are three separate embodiments of present invention as follows:

[A] A powder Raney catalyst obtained by

(i) melting nickel and aluminum,

(ii) quenching droplets of said melted mixture by means of dropping them onto chilled water through a nozzle to obtain a quenched lump alloy,

(iii) optionally breaking the quenched lump alloy,

(iv) classifying and activating the alloy of step (ii) or (iii),

(v) using said alloy of step (iv) as a Raney catalyst in a hydrogenation reaction,

(vi) collecting said alloy of step (v),

(vii) crushing said Raney catalyst used in the hydrogenation reaction into powder, and

(viii) reactivating.

[B] A powder Raney catalyst obtained by

(i) melting nickel and aluminum,

(ii) quenching droplets of said melted mixture by means of dropping them onto chilled water through a nozzle to obtain a quenched lump alloy,

(iii) optionally breaking the quenched lump alloy,
(iv) classifying and activating the alloy of step (ii) or
(iii),
(v) using said alloy of step (iv) as a Raney catalyst in a
hydrogenation reaction,
(vi) collecting said alloy of step (v),
(vii) crushing said Raney catalyst used in the hydrogenation
reaction into powder, and
(viii) reactivating; and
a step of adding molybdenum and/or tin up to 15%, wherein at
least one of molybdenum and tin must be in the catalyst.

[C] A powder Raney catalyst consisting of a nickel aluminum
alloy with molybdenum and/or tin up to 15% made by the process
comprising

melting a mixture of nickel and aluminum,
quenching droplets of said melted mixture of nickel and
aluminum by means of dropping them onto chilled water though a
nozzle to form a quenched lump alloy which has the grain diameter
of 1mm to 15mm,
activating said quenched lump alloy particles to form a Raney
catalyst, and
crushing said Raney catalyst to form a powder.

REFERENCES RELIED UPON IN THE FINAL REJECTION

The final Office Action dated December 18, 2001, relies on the following references:

- (1) Diffenbach et al., US 3,719,732;
- (2) Lepper et al., US 4,520,211;
- (3) Schuetz et al., US 5,536,694;
- (4) Raney US 1,628,190; and
- (5) Richter US 3,673,116.

ISSUES ON APPEAL

[1] Whether claims 1, 18 and 20 should stand rejected under 35 U.S.C. §103(a) as being unpatentable over Diffenbach et al. in view of Lepper et al.

[2] Whether claims 1 and 20 should stand rejected under 35 U.S.C. §102(e) as anticipated by the Prior Art Section of Schuetz et al.

[3] Whether claims 1 and 20 should stand rejected under 35 U.S.C. §103(a) as obvious over the Prior Art Section of Schuetz et al.

[4] Whether claims 1, 18 and 20 should stand rejected under 35 U.S.C. §103(a) as being unpatentable over the Prior Art Section of Schuetz et al. in view of Raney.

[5] Whether claims 1, 18 and 20 should stand rejected under 35 U.S.C. §103(a) as being unpatentable over the Prior Art Section of Schuetz et al. in view of Raney, Richter and Lepper et al.

Appellants want to make perfectly clear that during prosecution of this case, the Examiner has applied Schuetz et al. in a very unorthodox manner. The Examiner views Schuetz et al. as containing two separate disclosures, i.e., i) the Detailed Description of the Invention Section, and ii) the Prior Art Section. Based on Appellants' September 27, 2001 Amendment, ~~the Examiner has withdrawn the rejections based on the Detailed Description of the Invention Section of Schuetz et al.~~; however, the Examiner has maintained the rejections based on the Prior Art Section of Schuetz et al. See the paragraph bridging pages 9-10 of the December 18, 2001 Office Action.

In the above rejections numbered (2) - (5), Schuetz et al. has been cited for what is taught in the Prior Art Section only.

GROUPING OF CLAIMS

It is respectfully submitted that the presently appealed claims 1, 18 and 20 should each be considered to stand and fall separately. In other words Group [A] is claim 1, Group [B] is claim 18 and Group [C] is claim 20.

Appellants respectfully submit that claim 1 should be grouped differently than claim 18, since claim 18 requires at least one of molybdenum and tin, whereas claim 1 does not require at least one of the molybdenum and tin. In the rejections numbered [3] and [4] above, the Examiner has taken the position that claim 1 is rejected solely over the reference of Schuetz et al., whereas the Examiner requires the combination of Schuetz et al. and Raney in order to find claim 18 unpatentable. Accordingly, the presence of tin and/or molybdenum in the claimed catalyst, is a feature which differentiates the catalyst from Schuetz et al. and should be considered separately on appeal.

Appellants respectfully submit that claims 1 and 20 should be given a separate grouping, since claim 20 recites that the quenched lump alloy has a grain diameter of 1 mm to 15 mm, whereas claim 1 does not have a limitation of the size of the quenched lump alloy. As noted in the paragraph bridging pages 9-10 of the December 18, 2001 Final Office Action, the Examiner is relying solely upon the Prior Art Section of Schuetz et al. in finding claims 1 and 20 obvious in the rejection numbered as [3] above. It is clear that the Prior Art Section of Schuetz et al. is silent with respect to the size of the quenched catalyst droplets. Accordingly, the catalyst having a specific particle diameter range in claim 20 should be considered separately from the

catalyst of claim 1 such that both should be considered separately.

Appellants respectfully submit that claim 18 should be grouped separately from claim 20, since the catalyst of claim 18 must contain molybdenum and/or tin, whereas the catalyst of claim 20 does not. In the rejection numbered as [3] above, the Examiner rejects claims 1 and 20 over the Prior Art Section of Schuetz et al., and in the rejection numbered as [4] above, the Examiner rejects claims 1, 18 and 20 over the combination of the Prior Art Section of Schuetz et al. and Raney. Thus, the Examiner must be relying on Raney to teach that the catalyst may contain molybdenum and/or tin. This distinction leads Appellants to reasonably conclude that the recitation of molybdenum and/or tin in claim 18 is a feature which is of sufficient weight to keep claims 18 and 20 in separate groups.

Below is a summary of the arguments indicated in the Final Office Action by the U.S. Examiner, followed by the arguments of Appellants in support of the patentability of the appealed claims. In the Examiner's arguments, the Examiner stresses that certain references teach that the catalyst can be used in the particle form or the powder form for the hydrogenation of sugars to form sugar alcohols. However, the cited references, in any combination, fail to teach or suggest taking a lump form catalyst which has

been used in a hydrogenation reaction, crushing it to a powder and reactivating the powder catalyst, as presently claimed.

SUMMARY OF EXAMINER'S ARGUMENTS REGARDING REJECTION [1]

In support of the rejection of claims 1, 18 and 20 under 35 U.S.C. §103(a) as being unpatentable over Diffenbach et al. in view of Lepper et al., the Final Office Action of December 18, 2001 included the following arguments:

Diffenbach '732 discloses a process for producing active catalyst particles as stated in the above rejection. Diffenbach '732 teaches that the product can be conveniently used as fixed bed catalyst in contrast to the commonly used powdered catalyst which require the addition of a filtering step for removal from a product stream (note column 1, lines 53-65).

The difference is Diffenbach '732 does not disclose the step of crushing the catalyst particles.

Lepper '211 teaches that in a process of making polyhydric alcohols (i.e. sugar alcohols) by the hydrogenation of carbohydrates, a "catalyst solid bed" is preferred. The catalyst solid bed is defined as stationary arrangement of the catalyst in the reactor in the manner of a packed bed (note column 2, lines 47-52), thus the catalyst solid bed in Lepper is considered the same as a fixed bed catalyst. Lepper further discloses that the particle size of the catalyst employed in lumpy form may vary widely. On the one hand, the catalyst particles should not be so small that the flow resistance of the catalyst solid bed greatly hinders the through flow of the mixture of hydrogen and aqueous carbohydrate solution and necessitates too high a pressure. On the other hand, the maximum dimensions of the lumpy catalyst are given by the reactor geometry. As a rule, the particle size of the catalyst lumps, i.e. the diameter and/or length of the catalyst particles, will be selected so that it is in the range of from about 2 to 10 mm (note column 3, lines 16-32). This range overlaps the claimed range. The subject matter as a whole would have been obvious to one having ordinary skill in the art at the time the invention was made to have selected the overlapping portion of the range

disclosed by the reference because overlapping ranges have been held to be a *prima facie* case of obviousness, see *In re Malagari*, 182 U.S.P.Q. 549. It is noted that the in *Lepper '211*, the catalyst is a ruthenium-containing catalyst, not a Raney catalyst, however, only the physical size of the catalyst (not the composition of the catalyst) has direct impact on the flow resistance, etc., as discussed above, for the hydrogenation process.

It would have been obvious to one of ordinary skill in the art at the time of the invention was made to slightly reduce the particle size of the product of *Diffenbach* in order to obtain a more desirable particle size range for a fixed bed, as suggested by *Lepper '211*.

APPELLANTS ARGUMENTS SUPPORTING PATENTABILITY OVER DIFFENBACH ET AL.

AL. AND LEPPER ET AL.

In response to the above-reasons indicated for the rejections under 35 U.S.C. § 103 in the Final Office Action, the following arguments are respectfully submitted in support of the patentability of the appealed claims.

Present Invention and Its Advantages

The present invention is drawn to:

a powder Raney catalyst obtained by

(i) melting nickel and aluminum,

(ii) quenching droplets of said melted mixture by means of dropping them onto chilled water through a nozzle to obtain a quenched lump alloy,

(iii) optionally breaking the quenched lump alloy,

- (iv) classifying and activating the alloy of step (ii) or (iii),
- (v) using said alloy of step (iv) as a Raney catalyst in a hydrogenation reaction,
- (vi) collecting said alloy of step (v),
- (vii) crushing said Raney catalyst used in the hydrogenation reaction into powder, optionally
- (viii) reactivating the powdered catalyst, and optionally a step of adding molybdenum and/or tin to the catalyst.

In other words, the present invention relates to recyclable lump form Raney catalyst and to powder type Raney catalyst obtained by reactivating used lump form Raney catalyst.

The present invention is characterized in that the powder type Raney catalyst is consistently obtained as a highly active powder type Raney catalyst by reusing used lump form Raney catalyst no matter how many hydrogenation cycles it has been exposed to. In other words, used lump form Raney catalyst can be reused as raw material for powder type Raney catalyst whether the catalytic activity of the used lump form catalyst remains or not. Moreover, the thus obtained powder type Raney catalyst is available with as high catalytic activity as the conventional Raney catalyst has, even though the powder type catalyst has been obtained from the used catalyst.

A cross-sectional view of an activated lump form Raney catalyst of the present invention has been enlarged by a scanning electron microscope (PHOTO 1, see the attached).

This photograph is evidence that the activated lump form Raney catalyst has different layers. The outer layer which appears bright in the photograph represents an activated catalyst whereas the inner layer which appears relatively dark represents the inactive form of the catalyst. Thus, there is a large amount of lumped inactivated catalytic alloy (nickel-aluminum alloy) inside the activated catalytic layer. The inactive layer is possible, since the alkali solvent, such as an aqueous sodium hydroxide solution, is prevented from penetrating into the core of the particle by the active surface layer.

The lump form Raney catalyst which has an evenly thick active surface layer of the catalytic alloy will lose its activity upon repeated use as a hydrogenation catalyst. However, the catalytic alloy existing inside of the used lump form Raney catalyst can be activated by crushing the lump catalyst, thereby exposing the inactive inner layer followed by activation treatment with an alkali solution.

Distinctions Between the Presently Claimed Invention of
Claims 1, 18 and 20 and the Teachings of Diffenbach et al. and
Lepper et al.

First, Appellants will show that Diffenbach et al. fail to teach or suggest: (A) a step of quenching the molten nickel and aluminum into a quenched **lump** alloy; and (B) a step of crushing the Raney catalyst into a powder. Second, Appellants will show that Lepper et al. fail to cure the deficiencies of Diffenbach et al.

Diffenbach et al. is clearly concerned with the preparation of a high surface area catalyst, see Figures 1, 4 and 5 of Diffenbach et al., and Diffenbach et al. fail to teach or suggest: (A) a step of quenching the molten nickel and aluminum into a quenched **lump** alloy; and (B) a step of crushing the Raney catalyst into a powder. The inventive catalyst is different in shape from that of Diffenbach et al., possibly due to the temperature of molten alloy which is subjected to the quenching step i.e. the melting point of the molten alloy.

An Al-Ni phase diagram (Teruo KUBOMATSU, Shinichiro KOMATSU, Raney catalyst, page 27, May 10, 1971) is attached hereto for reference. The diagram shows that the melting point of a melted aluminum-nickel mixture and the composition of an alloy obtained in quenching the molten metal of the composition.

The Al-Ni phase diagram is commonly used to show the melting point of an Al-Ni alloy and the composition of the alloy obtained upon cooling. Its vertical axis represents the temperature, while the lower horizontal axis represents an atomic (mole) ratio of Ni and Al in the mixture and the upper horizontal axis indicates a weight ratio of Ni and Al in the mixture. The uppermost curve shows the temperature where the mixture of the composition ratio is completely molten i.e. the melting point of the alloyed substance. The portion under the curve or at lower temperatures represents the solid portion, showing the metallic composition of solidified Al-Ni alloys. For example, an alloy having a composition of NiAl is dominant in cooling at room temperature a molten metal having the range of 45 to 59 atomic% Ni. On the other hand, an alloy of composition of Ni_2Al_3 dominates in the range of 36 to 41 atomic% Ni. Two types of alloys of compositions Ni_2Al_3 and NiAl coexist in the range of 41 to 45 atomic% Ni at the interval between the above.

The difference between the present invention and the catalyst disclosed by Diffenbach will be explained here below, referring to the phase diagram.

The catalyst disclosed by Diffenbach has a high aluminum content. All catalytic alloys produced in Diffenbach's Examples have an aluminum content of 70 weight % or more. When the aluminum content is 70 weight % (i.e. nickel content of 30 weight %), the

melting point is about 900°C according to the attached Al-Ni phase diagram. Therefore, the melting point of catalytic alloy produced in Diffenbach's Examples does not exceed 900°C at a maximum.

When an alloy molten in the above temperature range is dripped into water, something like a water vapor film is thought to be formed by evaporation of water on the surface of the molten metal. However, the water vapor film formed at this temperature has such a poor strength that the ambient cooling water breaks the water vapor film, likely to have a direct contact with the molten metal. Then, the cooling water is thought to come into direct contact with the catalyst, resulting in a minute water vapor explosion to form a catalyst shaped as described by Diffenbach in the specification.

On the other hand, the aluminum content is lower in the present invention than disclosed by Diffenbach (usually 66.6 to 33.3 weight % or preferably 50 weight %), having a nickel content of 33.3 to 66.6 weight % or preferably 50 weight %. The melting point of the alloy then is about 950°C at a nickel content of 33.3 weight %, about 1300°C at a nickel content of 50 weight %, and about 1600°C at a nickel content of 66.6 weight % according to the Al-Ni phase diagram. In other words, the melting point of the catalytic alloy of the present invention is at least 950°C and more preferably as high as 1300°C. This is much higher than the catalyst of Diffenbach.

When molten catalytic alloy having such a high melting point is dripped into water, water evaporates on the surface of molten alloy as in the case of Diffenbach. However, water vapor is thought to be instantaneously generated by the high temperature of the molten alloy, thereby forming a stronger water vapor film on the catalytic surface. Then, this high strength water vapor film formed there is thought to prevent water vapor explosion on the catalytic surface due to direct contact between the catalytic alloy and the cooling water. As a result, the catalytic alloy of the present invention is thought to be solidified in lump as dripped, instead of being formed as described by Diffenbach.

As explained above, the catalysts of Diffenbach are clearly different in melting temperature from the catalysts of the present invention, and this difference in temperature is thought to influence the shapes of catalysts obtained by quenching the molten alloy such that the catalyst particles are not in the lump form as recited in the present claims.

The following explanation is provided to show that there are additional distinctions between the present invention from the teachings of Diffenbach et al.

Diffenbach et al. describe their own invention in column 2, lines 40 to 51, as follows:

"Broadly, this invention comprises methods of forming aluminum-nickel, aluminum-cobalt, aluminum-copper and

aluminum-iron alloys, or mixtures, into shaped particles of comparatively high surface area. This property of high surface area is of concern, since the preferred end use for these shaped particles is in the formation of highly active metal catalysts of the Raney type. As used in this application, a Raney nickel, cobalt, copper or iron has been put in a high surface area, highly active condition via the technique of alkali leaching the aluminum content out of the alloy."

As seen from this description, Diffenbach et al. teach that a high surface area of the catalyst is an important factor to obtain a highly active catalyst.

Then, Diffenbach et al. refer to various cooling methods used in production of Raney catalyst to obtain grained molded active catalyst from molten alloy. Diffenbach et al. use water as a cooling means for molten alloy, describing the shape of molded molten alloy quenched by the use of water in column 3, lines 27 to 47, as follows:

"The basic technique most useful for alloy shaping is melt forming, that is, putting the alloy into a shape while in a melt condition with subsequent cooling to ambient temperatures. The shapes of preferred importance are those of FIGS. 4 and 5, although essentially any shape can be utilized. The aim of any shape, however, is to attain a high surface area while maintaining crush resistance and still minimizing pressure drop on the fixed bed reactors. The shapes of this invention are produced by dropping molten alloy into or onto a cooled medium. When dropped onto a cold medium such as a plate, the drop will spread to the form of a disc. If a shaped cold surface is used, the drop will form a shape complementary to the cold surface to which it was contacted. When dropped into a cold medium, which can be water or some surface, the drop deforms to form a novel shape depending on the liquid, the energy of the falling

drop, and its melt condition. The preferred particle shape from this technique is an essentially hollow, partially spherical particle. Particles of any of these types have proven to subsequently yield very active Raney catalyst."

The specification of Diffenbach et al. says that "essentially any shape can be utilized." (see column 3, lines 31-32) as the shape of molded catalyst disclosed by Diffenbach et al., while referring to a higher catalytic surface area as a preferred embodiment. Moreover, it discloses "an essentially hollow, partially spherical particle" (see column 3, lines 45-46) by way of concrete example of its preferred particle shape. These descriptions are evidence that the catalyst disclosed by Diffenbach et al. is based on a technical idea quite distinct from that of the present invention. The invention of Diffenbach et al. aims to obtain the catalyst having a high activity by means of increasing the surface area of the catalyst by forming the grained catalyst into a concave shape. While the present invention is characterized in that the lump form catalytic alloy is activated only on the surface layer, keeping inactive catalytic alloy inside the catalyst, thereby enabling it to be reused into powder type Raney catalyst even after rendered inactive as the lump form Raney catalyst through repeated hydrogenation.

The concept of reusing a catalyst by the method of the present invention is obviously missing in the catalyst of Diffenbach et al. constructed to have a cavity inside, having no

catalytic alloy. On the contrary, the present invention can not be realized by using a catalyst shaped as disclosed by Diffenbach et al., since no catalytic alloy for reactivation remains inside lump form Raney catalyst.

Also, Appellants respectfully submit that Diffenbach et al. teach away from using a powder catalyst, see column 2, line 57 to column 3, line 35. Diffenbach et al. state that it is an object of their invention to shape the catalyst prior to activation. The type of alloy is chosen to minimize the possibility that the catalyst particles "will crush to a powder consistency... and that the particles would not be entrained in the product stream."

A reference which leads one of ordinary skill in the art away from the claimed invention cannot render it unpatentably obvious. *Dow Chem. Co. v. American Cyanamid Co.* 816 F2d 617, (CAFC 1987). In determining the scope and content of the prior art, and determining whether the prior art suggested the claimed invention, the references "must be read as a whole and consideration must be given where the references diverge and teach away from the claimed invention." *Akzo N.V. v. United States Int'l Trade Comm'n*, 1 USPQ2d 1241, 1246 (Fed. Cir. 1986); *In re Fine*, 5 USPQ2d 1596, 1598-99 (Fed. Cir. 1988).

Thus, significant patentable distinctions exist between the present process and the process of Diffenbach et al.

The Examiner, aware of the deficiencies of Diffenbach et al. cites Lepper et al. in order to cure the deficiencies of Diffenbach et al. Appellants respectfully submit that Lepper et al. fail to cure the deficiencies of Diffenbach et al.

Appellants note from the paragraph bridging pages 5-6 of the December 18, 2001 Office Action, that the Examiner has cited Lepper et al. for teaching that the "particle size of the catalyst employed in lumpy form may vary widely."

Appellants respectfully submit that Lepper et al. teach away from using a catalyst in the powder form. In column 3, lines 16-32, Lepper et al. teach that the "catalyst particles should not be so small that the flow resistance of the catalysts solid bed greatly hinders the through-flow of the charged mixture of hydrogen and aqueous carbohydrate solution and necessitates to high a pressure."

As mentioned above, *Dow Chem. Co. v. American Cyanamid Co.* stands for the fact that a reference which leads one of ordinary skill in the art away from the claimed invention cannot render it unpatentably obvious.

In addition, Lepper discloses lump catalyst in which metal ruthenium is carried on a carrier material such as activated carbon. On the other hand, the product of the present invention is made from lump Raney catalyst obtained from metal molten and quenched in water. In other words, the inventive catalyst

essentially consists only of alloy components, and does not contain a "carrier" which gives no direct contribution to the hydrogenation reaction.

Clearly the skilled artisan would not be motivated to look to the teachings of Lepper regarding the catalyst particle size to modify the Raney catalyst of Diffenbach et al., since Lepper's catalyst is chemically/physically bound to the support. The artisan would reasonably conclude that any advantages associated with the particle size of Lepper's catalyst are linked to the fact that the catalyst particles are on a support.

Even assuming *arguendo* that the catalyst as disclosed by Lepper could be once used and then, milled to get powder Raney catalyst as in the present invention, in such a case, the catalyst contains a "carrier" which gives no direct contribution to the hydrogenation reaction. Therefore, the thus obtained powder Raney catalyst is **obviously poorer** in hydrogenating efficiency than that of the present invention.

Lepper et al. fail to teach or suggest the advantages of a crushing step wherein the Raney catalyst is crushed to form a powder as required by inventive claim 1 and 20.

Accordingly, the combination of Diffenbach et al. and Lepper et al. fail to teach or suggest (A) a step of quenching the molten nickel and aluminum into a quenched **lump** alloy; and (B) a step of crushing the Raney catalyst into a powder. According to the MPEP,

a *prima facie* case of obviousness can not be said to exist unless all of the limitations of the presently claimed invention are either taught or fairly suggested. See MPEP § 2143.03. As such, significant patentable distinctions exist between the present invention and the teachings of Difffenbach et al. and Lepper et al.

SUMMARY OF EXAMINER'S ARGUMENTS REGARDING REJECTIONS [2] AND [3]

In support of the rejection of claims 1 and 20 under 35 U.S.C. §102(e) as anticipated by, or in the alternative under 35 U.S.C. §103(a) as being unpatentable over the Prior Art Section of Schuetz et al., the Final Office Action of December 18, 2001 included the following arguments:

Schuetz '694 discloses that activated metal catalysts are known as Raney catalysts in the chemical engineering field. They are used mainly in the powdered form in a large number of reactions for hydrogenating organic compounds. These powdered catalysts are prepared from an alloy of a catalytically active metal and another alloying component which is leachable in alkalis. The Raney process metals used are mainly Ni, Co, Cu or Fe. The alloying component which is mainly used is Al (note column 1, lines 30-41). This so called Raney alloy is first finely milled according to Raney's method. Then the aluminum is completely or partially removed by leaching with alkalis such as, for example, caustic soda solution (note column 1, lines 42-50).

The powder Raney catalyst as disclosed in Schuetz '694 anticipates the claimed product.

Alternatively, any difference imparted by the product by process limitations would have been obvious to one having ordinary skill in the art at the time the invention was made because where the examiner has found a substantially similar product as in the applied prior art the burden of proof is shifted to the applicant to establish that their product is patentably distinct not the examiner to show that the same process of making,

see *In re Brown*, 173 U.S.P.Q. 685, and *In re Fessmann*, 180 U.S.P.Q. 324.

APPELLANTS ARGUMENTS SUPPORTING PATENTABILITY OVER THE PRIOR ART

SECTION OF SCHUETZ ET AL.

In response to the above-reasons indicated for the rejections under 35 U.S.C. §§ 102 and 103 in the Final Office Action, the following arguments are respectfully submitted in support of the patentability of the appealed claims.

In the Prior Art Section of Schuetz et al., Schuetz et al. characterize the state of the prior art as teaching that Raney alloys are available as a "coarse particulate," and that the activation of the particulate takes place only in a "shallow surface layer whose thickness can be set by means of the extraction conditions." See column 1, lines 55-63.

According to Schuetz et al.:

[t]he major disadvantage of catalysts prepared using this method is the poor mechanical stability of the relatively thin activated outer layer. Since only this outer layer of the catalyst is also catalytically active, this results in rapid deactivation which at best can be partially reversed by renewed activation of deeper-lying alloyed layers using caustic soda solution. (See column 1, line 64 to column 2, line 3). (Emphasis added).

Thus, the Prior Art Section of Schuetz et al. teach that at best, the used lump alloy containing an inactive outer coating can be partially reactivated chemically with a caustic soda solution.

Schuetz et al. fail to teach or suggest the inventive step (vii) of crushing said Raney catalyst used in the hydrogenation reaction into powder to thereby renew activation of the deeper-lying alloyed layers. As the MPEP directs, all the claim limitations must be taught or suggested by the prior art to establish a *prima facie* case of anticipation or obviousness. See MPEP §§ 2131 and 2143.03.

Accordingly, significant patentable distinctions exist between the process of present claims 1 and 20 and the Prior Art Section of Schuetz et al.

SUMMARY OF EXAMINER'S ARGUMENTS REGARDING REJECTION [4] AND [5]

In support of the rejection of claims 1, 18 and 20 under 35 U.S.C. §103(a) as being unpatentable over the Prior Art Section of Schuetz et al., in view of Raney and optionally further in view of Richter and Lepper, the Final Office Action of December 18, 2001 included the following arguments:

Schuetz '694 discloses that activated metal catalysts are known as Raney catalysts in the chemical engineering field, they are used mainly in the powdered form in a large number of reactions for hydrogenating organic compounds. These powdered catalysts are prepared from an alloy of a catalytically active metal and another alloying component which is leachable in alkali. The catalytically active metals used are mainly Ni, Co, Cu or Fe. The alloying component which is mainly used is aluminum (note column 1, lines 30-41). Schuetz '694 further teaches that powder catalysts have the disadvantages that they can only be used in batch processes and have to be isolated after the catalytic reaction by time-consuming filtration of the reaction

media. Various processes for preparing molded items have therefore been disclosed which lead to activated metal fixed bed catalysts after extraction of the aluminum. Thus, for example, coarse particulate, i.e. only coarsely milled, Raney alloys are available which can be activated by treatment with caustic soda solution (note column 1, lines 51-60). When coarse particulate is desired to be used in fixed bed catalyst, it would have been obvious to one of ordinary skill to remove any particles with undesirable size before activating the Raney alloy.

Thus, Schuetz '694 fairly teaches that both the powder form and the coarse particulate form of Raney catalyst are known in the art. They can be formed by finely or coarsely milled Raney alloys. Thus, it would have been obvious to one of ordinary skill in the art at the time of the invention was made to further pulverize the coarse particles to obtain finer particles when powder catalyst is desired. It should be noted that in Schuetz '694, both powder catalyst or coarse catalyst can be used.

For claims 1, 5 and 20, "up to 15%" of Mo and/or Sn includes zero. For claims 18, 19, Schuetz '694 discloses that it is known in the art to include promoters, such as chromium, iron, cobalt, tantalum, titanium and/or molybdenum and also metals from the platinum group, up to 15%, in the Raney catalysts to influence their catalytic properties (note paragraph bridging columns 5-6).

The difference is Schuetz '694 does not specifically disclose the process of making the Raney alloys even though Schuetz '694 does disclose that such alloys are known in the art.

Raney '190 discloses a method of producing metallic nickel in a catalytic state such as may be used in the hydrogenation of oils, fats, waxes and the like (note page 1, lines 5-7). The process comprises the steps of alloying metallic nickel with metals such as silicon and aluminum in various proportions, and then dissolving the aluminum and silicon from the alloy by means of a solvent which will not attack the nickel, whereupon the nickel remains in a finely divided state (note page 1, lines 8-13). The alloying is carried out by melting the nickel, aluminum and silicon either separately, or together, cooling the melt and pulverizing the solidified alloy (note page 1, lines 32-34). Raney '190 further discloses that the nickel aluminum alloy may be either very finely pulverized or it may be broken in

pieces the size of peas or smaller. In either condition, the alloy may be treated with caustic soda or the aluminum removed with the use of some other solvent. In case the larger pieces are used, the nickel is left in a more or less spongy and porous state, somewhat similar to a cinder, and for certain classes of work is necessary and desirable to have the catalyster in this condition (note page 1, lines 81-92). Raney '190 further discloses that the alloy can have 10-85% nickel and 90-15% aluminum (note page 1, lines 74-77). This range would create a Ni:Al weight ratio that overlaps the claimed range. The subject matter as a whole would have been obvious to one having ordinary skill in the art at the time the invention was made to have selected the overlapping portion of the range disclosed by the reference because overlapping ranges have been held to be a *prima facie* case of obviousness, see *In re Malagari*, 182 U.S.P.Q. 549.

Optionally, Richter '116 can be applied to teach in the well known process of making Raney catalyst, "cooling" is quenching, or at least cooling is preferred to be quenching, note in Example 1, "cooled" is used and in Example 2, "quenching" is used. Also, Richter discloses that it was found beneficial to cool rapidly the Raney alloy produced by melting by excluding air, since the fine crystalline texture of the Raney alloy which occurs during the quenching of the melt, is easy to homogenize (note column 2, lines 39-43).

It would have been obvious to one of ordinary skill in the art at the time of the invention was made to use the known Raney process such as the process disclosed in Raney '190 which comprises the steps of melting, cooling or quenching, pulverizing, to produce the Raney alloy in the process of Schuetz '694 and to only coarsely pulverize the alloy in order to use coarse particles in a fixed bed as disclosed in Schuetz '694.

Optionally, Lepper '211 can be applied as stated in the above rejection to teach the desired particle size for a fixed bed catalyst used in a hydrogenation process.

It would have been obvious to one of ordinary skill in the art at the time of the invention was made to coarsely milled the Raney alloy as disclosed in Schuetz '694 within the range suggested by Lepper '211 because such range is desired for a fixed bed catalyst in the hydrogenation process.

APPELLANTS ARGUMENTS SUPPORTING PATENTABILITY OVER THE PRIOR ARTSECTION OF SCHUETZ ET AL., RANEY, RICHTER AND LEPPER

In response to the above-reasons indicated for the rejections under 35 U.S.C. §103 in the Final Office Action, the following arguments are respectfully submitted in support of the patentability of the appealed claims.

The patentable distinctions between the presently claimed process and the teachings of the Prior Art Section of Schuetz et al., as described in the section above, is herein incorporated by reference.

The Examiner has taken the position that the Prior Art Section of Schuetz et al. fairly teach both the powder form and the coarse particulate form of the Raney catalyst and that it would be obvious to one of ordinary skill to pulverize the coarse particles to obtain finer particles when powder catalyst is desired.

Appellants take the position that Schuetz et al. are aware that the coarse catalyst particles which have been used in catalysis contain an inner layer which can be activated. However, contrary to the Examiner's position, Schuetz et al. teach that at best, the deeper-lying alloyed layers can be activated with additional caustic soda solution. There is no teaching or suggestion in Schuetz et al. to grind the coarse particles into

the powder form to access the deeper-lying alloyed layers for reactivation as asserted by the Examiner.

The Examiner, aware of the deficiencies of Prior Art Section of Schuetz et al., cites Raney to cure the deficiencies. Appellants respectfully submit that Raney fails to cure the deficiencies of Prior Art Section of Schuetz et al.

The present invention relates to a lump form Raney catalyst capable of being re-utilized to a powder catalyst and a powder Raney catalyst prepared from a used lump form Raney catalyst.

The Examiner has taken the position that Raney discloses: 1) that the metal is melted and cooled in order to obtain the Raney catalyst; and 2) that the Ni-Al alloy may be either very finely pulverized or it may be broken in pieces the size of peas or smaller and then activated by treatment with caustic soda.

Appellants respectfully submit that Raney fails to cure the deficiencies of the Prior Art Section of Schuetz et al., since Raney fails to teach or suggest that the used coarse particulate catalyst is capable of being crushed to expose inactive potential catalyst material which can then be activated for use in hydrogenating organic compounds, as presently claimed.

The Examiner, aware of the deficiencies of the combination of the Prior Art Section of Schuetz et al. and Raney, cites Richter to cure the deficiencies. Appellants respectfully submit that

Richter fails to cure the deficiencies of the combination of the Prior Art Section of Schuetz et al. and Raney.

Regarding independent claims 1 and 20, the Examiner cites Richter for teaching a cooling process of the melted alloy. Accordingly, Richter fails to cure the deficiencies of the combination of the Prior Art Section of Schuetz et al. and Raney, since Richter fails to teach or suggest that the used coarse particulate catalyst is capable of being crushed to expose inactive potential catalyst material which can then be activated for use in hydrogenating organic compounds, as presently claimed.

Appellants respectfully submit that inventive claim 18 is further distinguished from the teachings of Richter. The present catalyst contains a small amount of molybdenum and/or tin (see claims 18-19), and the alloy consists primarily of nickel and aluminum. This is in distinction to the catalyst of Richter which is a Raney alloy consisting of four elements including iron, titanium and zirconium. Thus, the composition of Richter is entirely different from the alloy of the present invention. It is known in the art that if the composition and ratio of metals in an alloy are changed, the physical properties, such as melting points, of the alloy can greatly differ. Thus, the skilled artisan would not find the teachings of Richter regarding the cooling process applicable to the catalyst of the Prior Art Section of Schuetz et al. and Raney.

Thus, the combination of the Prior Art Section of Schuetz et al., Raney and Richter teach catalysts which are quite distinct from the present catalyst. In addition, those skilled in the art would not find the inventive catalyst obvious, based upon the disclosures of these cited references.

The Examiner, aware of the deficiencies of the combination of the Prior Art Section of Schuetz et al., Raney and Richter cites Lepper et al. to cure the deficiencies. Appellants respectfully submit that Lepper et al. fail to cure the deficiencies of the combination of the Prior Art Section of Schuetz et al., Raney and Richter.

Appellants note from the paragraph bridging pages 5-6 of the December 18, 2001 Office Action, that the Examiner has cited Lepper et al. for teaching that the "particle size of the catalyst employed in lumpy form may vary widely."

Appellants respectfully submit that Lepper et al. teach away from using a catalyst in the powder form. In column 3, lines 16-32, Lepper et al. teach that the "catalyst particles should not be so small that the flow resistance of the catalysts solid bed greatly hinders the through-flow of the charged mixture of hydrogen and aqueous carbohydrate solution and necessitates to high a pressure."

As mentioned above, *Dow Chem. Co. v. American Cyanamid Co.* stands for the fact that a reference which leads one of ordinary

skill in the art away from the claimed invention cannot render it unpatentably obvious.

In addition, Lepper et al. disclose lump catalyst in which metal ruthenium is carried on a carrier material such as activated carbon. On the other hand, the product of the present invention is made from lump Raney catalyst obtained from metal molten and quenched in water. In other words, the inventive catalyst essentially consists only of alloy components, and does not contain a "carrier" which gives no direct contribution to the hydrogenation reaction.

Clearly the skilled artisan would not be motivated to look to the teachings of Lepper et al. regarding the catalyst particle size to modify the Raney catalyst of the Prior Art Section of Schuetz et al., since Lepper et al.'s catalyst is chemically/physically bound to the support. The artisan would reasonably conclude that any advantages associated with the particle size of Lepper et al.'s catalyst are linked to the fact that the catalyst particles are on a support.

Even assuming *arguendo* that the catalyst as disclosed by Lepper et al. could be once used and then, milled to get powder Raney catalyst as in the present invention, in such a case, the catalyst contains a "carrier" which gives no direct contribution to the hydrogenation reaction. Therefore, the thus obtained powder Raney catalyst is obviously **poorer** in hydrogenating

efficiency than that of the present invention and the skilled artisan would not be motivated to include such a step in the process.

Lepper et al. fail to teach or suggest the advantages of a crushing step wherein the Raney catalyst is crushed to form a powder as required by inventive claim 1 and 20, and as such, Lepper et al. fail to cure the deficiencies of the combination of the Prior Art Section of Schuetz et al., Raney and Richter. According to the MPEP, a *prima facie* case of obviousness can not be said to exist unless all of the limitations of the presently claimed invention are either taught or fairly suggested. See MPEP § 2143.03.

In view of the foregoing, the presently claimed invention is not rendered unpatentable based on the teachings of the combination of the Prior Art Section of Schuetz et al., Raney, Richter and Lepper et al.

Conclusion

If any questions remain regarding the above matters, please contact Appellant's representative, Garth M. Dahlen, Ph.D. (Registry #43,575), in the Washington metropolitan area at the phone number listed below.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

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- 1) PHOTO 1
- 2) An Al-Ni phase diagram (Teruo KUBOMATSU, Shinichiro KOMATSU, Raney catalyst, page 27, May 10, 1971)
- 3) Claims on Appeal (Appendix A)

APPENDIX ACLAIMS ON APPEAL

Claims 1, 11-18 and 20 are pending and claims 11-17 have been withdrawn from consideration as being drawn to nonelected subject matter. Claims 1, 18 and 20 are appealed and are as follows:

1. (Three Times Amended) A powder Raney catalyst obtained by
 - (i) melting nickel and aluminum,
 - (ii) quenching droplets of said melted mixture by means of dropping them onto chilled water through a nozzle to obtain a quenched lump alloy,
 - (iii) optionally breaking the quenched lump alloy,
 - (iv) classifying and activating the alloy of step (ii) or (iii),
 - (v) using said alloy of step (iv) as a Raney catalyst in a hydrogenation reaction,
 - (vi) collecting said alloy of step (v),
 - (vii) crushing said Raney catalyst used in the hydrogenation reaction into powder, and
 - (viii) reactivating.

18. The powder type Raney catalyst as defined in claim 1, further comprising a step of adding molybdenum and/or tin up to 15%, wherein at least one of molybdenum and tin must be in the catalyst.

20. (Twice Amended) A powder Raney catalyst consisting of a nickel aluminum alloy with molybdenum and/or tin up to 15% made by the process comprising

melting a mixture of nickel and aluminum,
quenching droplets of said melted mixture of nickel and aluminum by means of dropping them onto chilled water though a nozzle to form a quenched lump alloy which has the grain diameter of 1mm to 15mm,

activating said quenched lump alloy particles to form a Raney catalyst, and

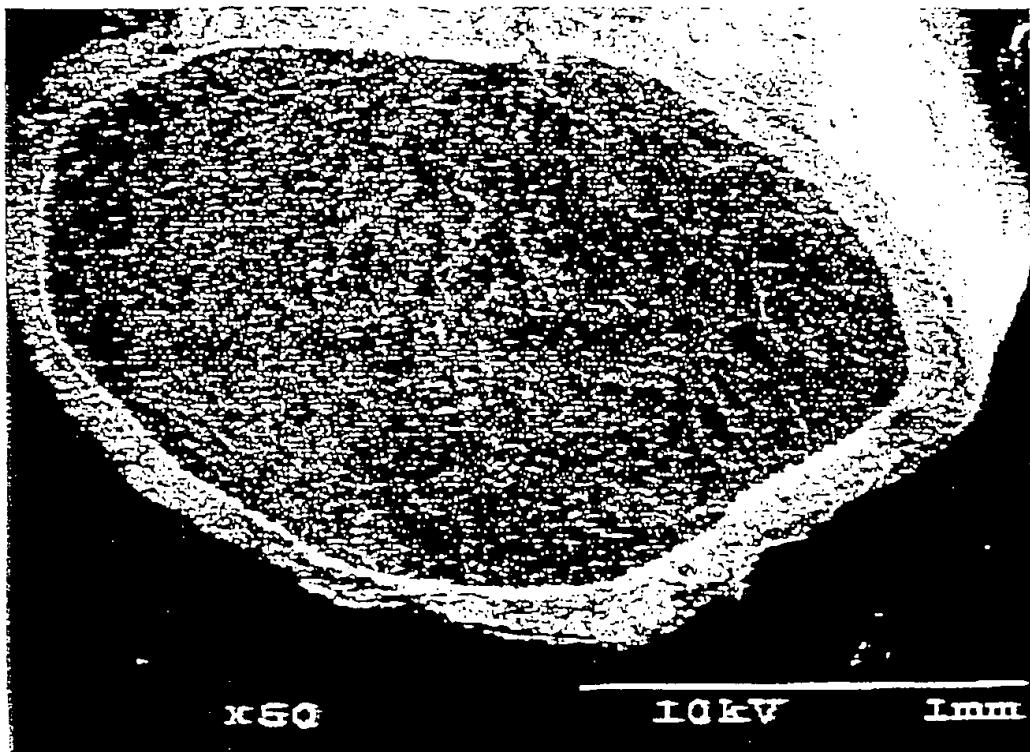
crushing said Raney catalyst to form a powder.

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SEM photography of lump form Raney catalyst of the present invention

Measuring device: HITACHI S-2460N

Enlargement ratio: 50 times



ラネー触媒

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2 合金の製造と粉碎

27

これらはあくまでも特種例である。電解はクニン酸1%, Na_2HPO_4 0.1%, $NaCl$ 0.5% を含む溶液中, 電圧4.2-4.7Vで行なった。

合金は普通微粉碎して使う。冷却条件は合金の粉碎の難易にも影響するが,

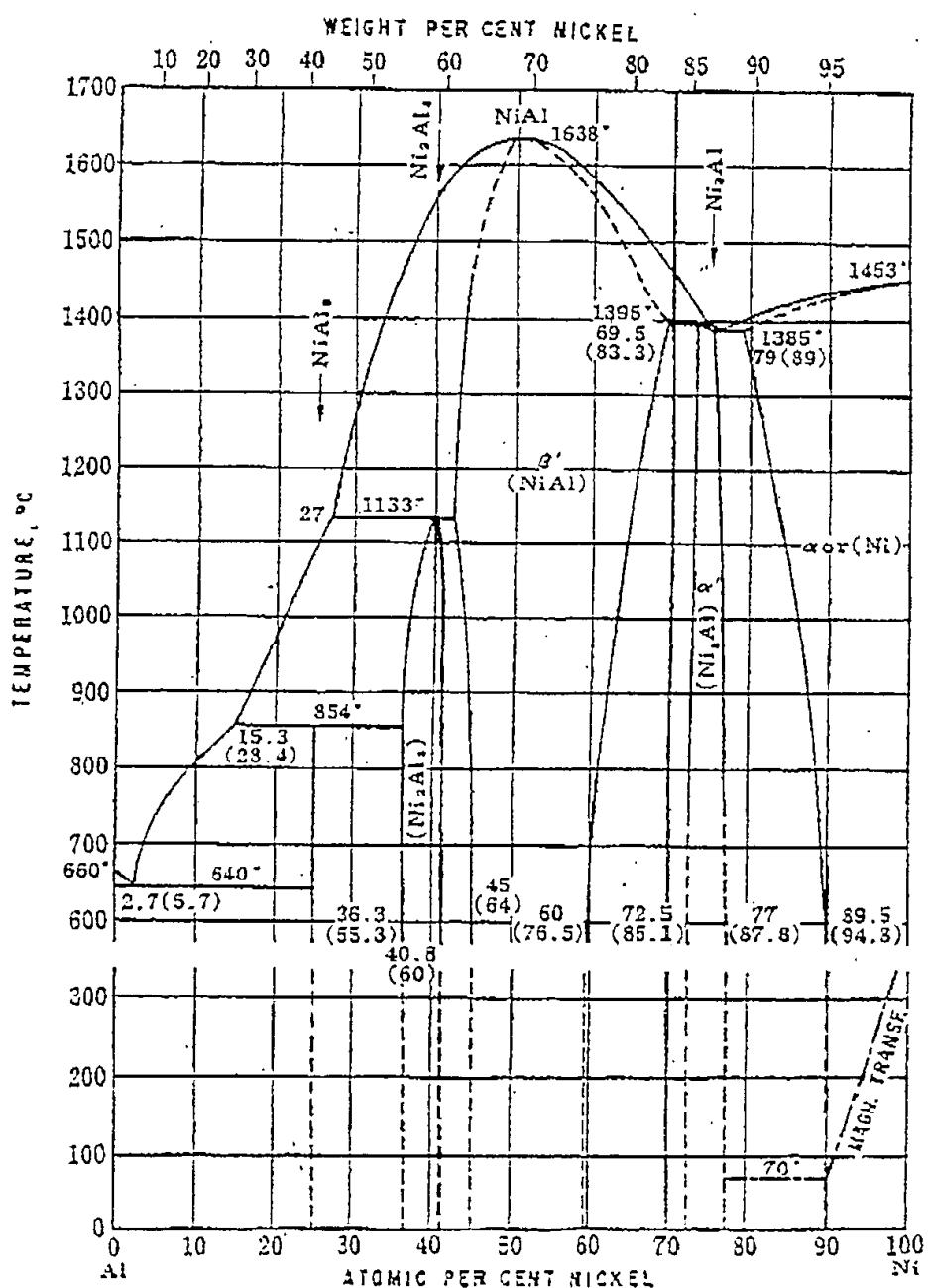


図 2.1 Ni-Al state diagram
by Max Hansen "Constitution of Binary alloys" 2nd Edition.

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